

Possible Three-Dimensional Nodes in the $s\pm$ Superconducting Gap of $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$

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We theoretically examine the superconducting state of $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$, an isovalent doping 122-iron-pnictide superconductor. We construct a three-dimensional ten-orbital model by first-principles band calculation, and investigate the superconducting gap within the spin-fluctuation-mediated pairing mechanism. The gap is basically $s\pm$, where the gap changes its sign between electron and hole Fermi surfaces, but three-dimensional nodal structures appear in the largely warped hole Fermi surface having a strong $Z^2/XZ/YZ$ orbital character. The present result, together with our previous study of the 1111 systems, explains the strong material dependence of the superconducting gap in iron pnictides.

KEYWORDS: 122 iron pnictides, superconducting gap, spin fluctuations, first-principles band calculation

It is now becoming clearer that the superconducting gap of iron pnictides¹ has nonuniversal forms. A number of experiments have suggested the presence of a fully open gap,^{2,3} but for LaFePO in particular, experiments have suggested the presence of line nodes in the gap.⁴⁻⁶ In a previous study, motivated by an experimental observation by Lee *et al.*,⁷ we have mainly focused on the 1111 systems, and pointed out that the band structure around the wave vector (π, π) in the unfolded Brillouin zone, and thus superconductivity, is sensitive to the pnictogen height measured from the iron plane.⁸ Using a five-orbital model,⁹ we have shown that when pnictogen is at high positions, the $X^2 - Y^2$ band around (π, π) gives a hole Fermi surface, and in this case, the spin fluctuations arising from the interaction between electron and hole Fermi surfaces give rise to a fully gapped $s\pm$ -wave pairing.⁹⁻¹² On the other hand, as the pnictogen is lowered, the Z^2 band around (π, π) rises up above the Fermi level, while the $X^2 - Y^2$ band sinks below. This results in the presence of line nodes in the superconducting gap either in the nodal $s\pm$ -wave form, where the gap nodes are on the electron Fermi surfaces, or in the d -wave form where the nodes are on the hole Fermi surfaces.^{9,13} Fluctuation exchange (FLEX)¹⁴ and functional renormalization group^{15,16} studies also found a similar tendency. The presence of line nodes in the superconducting gap results in a lower T_c , and this explains the experimental observation of line nodes in low- T_c LaFePO .⁴⁻⁶

However, if we focus on the 122 materials, there are now several experiments that do not fit into this view. In the isovalent doping system $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$,^{17,18} a number of experiments suggested the presence of line nodes in the superconducting gap,^{19,20} but T_c is relatively high (maximum T_c of about 30 K),¹⁸ in contrast to LaFePO . Moreover, the $X^2 - Y^2$ hole Fermi surface is found to be present in this material theoretically (as seen below), and this also seems to be supported by results of an angle resolved photoemission (ARPES) experiment.²¹ Another interesting observation in the 122 materials is the possible presence of nodes in the super-

conducting gap of KFe_2As_2 .²²⁻²⁶ In this material, the electron Fermi surface is barely present,²⁷ so it is likely that the gap nodes are on the hole Fermi surface. Here also, the $X^2 - Y^2$ hole Fermi surface should be present because there is a large number of holes. These experimental observations for the 122 materials do not seem to fit into the view that nodal pairing occurs when the $X^2 - Y^2$ hole Fermi surface disappears by reducing the pnictogen height. As for other origins of the presence of line nodes in the gap, the Coulomb avoidance,²⁸ as well as the competition of spin and orbital fluctuations,^{29,30} has been considered.

In the present study, we consider another possible origin of the gap nodes, which is peculiar to the 122 materials. We construct a *three-dimensional ten-orbital model* of $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ by first-principles calculation using maximally localized Wannier orbitals,³¹ and apply random phase approximation (RPA) to obtain the spin susceptibility and superconducting gap function. For the 122 materials, the Brillouin zone unfolding procedure⁹ that adopts the reduced unit cell (with one iron) cannot be performed strictly³² there fore, by adopting the ten-orbital model that uses the original unit cell (with two irons) of the body-centered tetragonal lattice structure, we fully take into account the peculiar features of the 122 band structure not present in the 1111 materials. The superconducting gap is basically $s\pm$, where the gap changes its sign between electron and hole Fermi surfaces, but when the hole Fermi surface around the Z point having a strong $3Z^2 - R^2$ (Z^2) orbital character becomes large by isovalent doping, the superconducting gap on that Fermi surface exhibits three-dimensional nodal structures. This kind of node is peculiar to the 122 systems, as also found in a five-orbital study of BaFe_2As_2 (but with different orbital characters of the Fermi surface).^{33,34} Since the Z^2 orbital does not play an important role in spin-fluctuation-mediated superconductivity, this explains why T_c in the isovalent doping system is relatively high despite the presence of nodes in the superconducting gap.

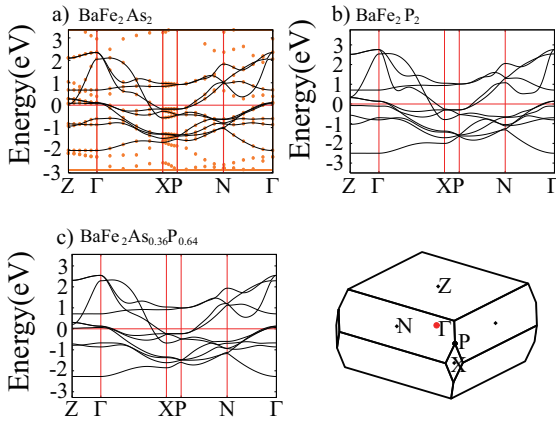


Fig. 1. (Color online) Band structures of the ten-orbital model of (a) BaFe_2As_2 , (b) BaFe_2P_2 , and (c) $\text{BaFe}_2\text{As}_{0.36}\text{P}_{0.64}$. In (a), the original first-principles band calculation is shown by dots. The Brillouin zone is shown in the inset.

In order to construct a realistic model, we first perform first-principles band calculation using the Quantum Espresso package³⁵ adopting the experimentally determined lattice structure,³⁶ and then obtain a ten-orbital model using maximally localized Wannier orbitals.³¹ There are ten bands mainly originating from the five 3d orbitals because there are two iron atoms per unit cell in the body-centered tetragonal lattice structure. We show in the upper panels of Fig. 1 the band structure of the ten-orbital models of BaFe_2As_2 and BaFe_2P_2 . To obtain a model of $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$, we first obtain a ten-orbital tight-binding model of “hypothetical” BaFe_2As_2 and BaFe_2P_2 having the experimentally determined lattice structure of $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ at each x . We assume that making a linear combination of the two sets of tight-binding parameters (hopping integrals and on-site energies) and mixing them with a ratio of $1-x : x$ will give a good approximation of the band structure of $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$. The band structure of this model with $x = 0.64$ is shown in the lower panel of Fig. 1.

In Fig. 2, we show the Fermi surfaces of $\text{BaFe}_2(\text{As}_{0.36}\text{P}_{0.64})_2$ for each orbital character, where the thickness represents the strength of the character. The 122 materials share common features with the 1111 materials in that they have three hole (around Γ -Z) and two electron Fermi (around X-P) surfaces. There are, however, some differences. One is that the portion of the Fermi surface around the Z point having a strong Z^2 character is continuously connected to the XZ/YZ portion of the Fermi surface around the Γ point in 122, while in 1111 the Z^2 Fermi surface, when present, is an isolated three-dimensional pocket. We will call this hole Fermi surface with a mixed Z^2 and XZ/YZ orbital character α_1 . Another difference of 122 from 1111 is that the hole Fermi surface having a strong $X^2 - Y^2$ orbital character coexists with the Z^2 Fermi surface; in 1111, either the three-dimensional Z^2 or the cylindrical $X^2 - Y^2$ (γ) Fermi surfaces exist depending on the pnictogen height.^{8,37-39} The hole Fermi surface with an $X^2 - Y^2$ character will be called γ , as in our study of

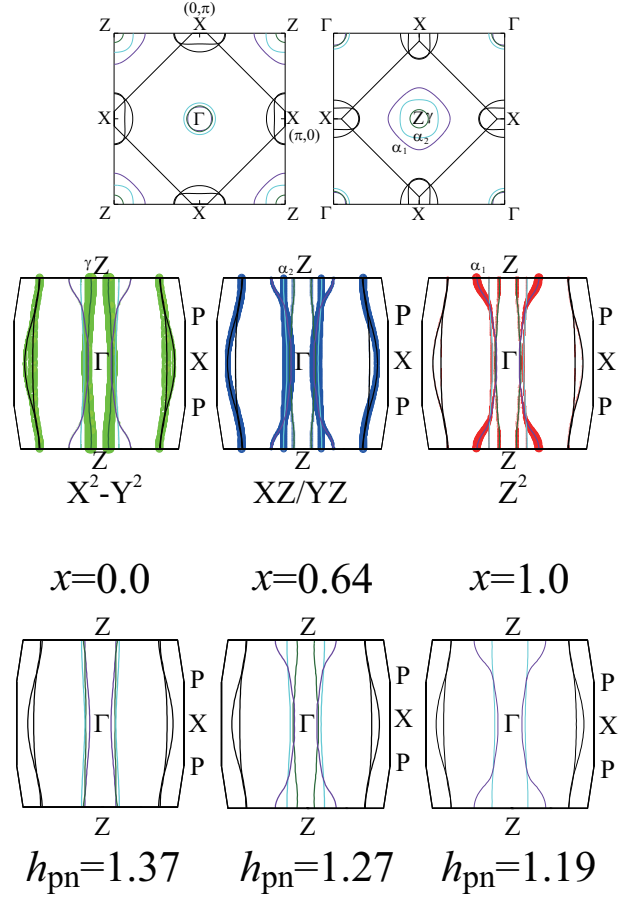


Fig. 2. (Color online) Upper panels: Fermi surfaces of the ten-orbital model of $\text{BaFe}_2(\text{As}_{0.36}\text{P}_{0.64})_2$. Horizontal (vertical) cuts are shown in the top (middle) panels. The vertical cuts are presented along with the strength of the orbital character. The X point corresponds to the wavevector $(\pi, 0)$ in the unfolded Brillouin zone. Bottom panels: evolution of the Fermi surface upon increasing the phosphorus concentration x . The pnictogen height h_{pn} is also presented.

1111.⁸ There is another hole Fermi surface having an XZ/YZ character, which we will call α_2 .

Replacing As by P does not alter the band filling (isovalent doping), and the main effect is to reduce the pnictogen height measured from the iron planes. In the bottom panels of Fig. 2, we show the vertical cut of the Fermi surface for various P contents. As in 1111, lowering the height leads to a higher Z^2 orbital level, resulting in a larger α_1 Fermi surface around the Z point.¹⁸ Such a strong warping of the Fermi surface has been directly observed in a recent ARPES experiment.⁴⁰ At the same time, the $X^2 - Y^2$ level is lowered, so that the γ Fermi surface shrinks, but it exists up to a P content of about $x \sim 0.7$ in the present calculation.

We now move on to the RPA calculation. We mainly concentrate on $\text{BaFe}_2(\text{As}_{0.36}\text{P}_{0.64})_2$ here, while we also comment on the calculation results for other x values. As for the electron-electron interaction, we consider the intra-orbital U , the interorbital U' , Hund's coupling J , and the pair hopping interaction J' . We consider the orbital-dependent interactions obtained in ref.⁴¹. We apply RPA in this model,^{8,9} and obtain the spin and charge suscep-

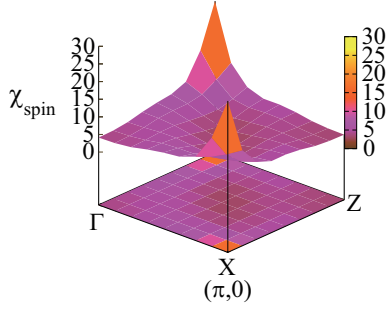


Fig. 3. (Color online) Largest eigenvalue of the spin susceptibility matrix for the ten-orbital model of $\text{BaFe}_2(\text{As}_{0.36}\text{P}_{0.64})_2$. The X point corresponds to the $(\pi, 0)$ point in the unfolded Brillouin zone.

tibility matrices. From these, we obtain the pairing interaction, which are plugged into the linearized Eliashberg equation. In the RPA calculation (where the self-energy correction is neglected), realistic values of the interaction results in very large spin fluctuations, so we multiply all the electron-electron interaction by a factor f .⁸ In this way, we keep the relative strength of the interactions between different orbitals to be the same as those obtained from the first-principles calculation. In the following, we will present the eigenfunction of the Eliashberg equation in the band representation, and call them the “(superconducting) gap”. In the actual calculation results shown below, we take $16 \times 16 \times 16$ k -point meshes, 128 Matsubara frequencies, $T = 0.07$ eV, and the interaction reducing ratio $f = 0.55$.

In Fig. 3, we show a plot of the eigenvalue of the spin susceptibility matrix. It has a peak at the X point (corresponding to the wave vector $(\pi, 0)$ in the unfolded Brillouin zone, although the unfolding cannot be strictly performed), which originates from the interaction between electron and hole Fermi surfaces. As shown in our earlier study,⁸ the main interaction that induces the spin fluctuation comes from the $X^2 - Y^2$ and XZ/YZ portions of the Fermi surface, namely, the intraorbital interaction within these orbitals. Performing a similar calculation for other values of x , we find that the spin susceptibility is suppressed monotonically upon increasing the phosphorus content, as expected from the fact that the γ Fermi surface shrinks.⁸

We show the superconducting gap in Fig. 4. The electron and hole Fermi surfaces have different signs of the gap for most portions, namely, the gap is basically \pm , originating from the spin fluctuation mentioned above. The gap on the electron Fermi surfaces is fully open since the $X^2 - Y^2$ γ Fermi surface is present, as is the case for the 1111 systems with high pnictogen positions.⁸ On the other hand, as the volume of the α_1 Fermi surface around the Z point grows upon lowering the pnictogen height by isovalent doping, a three-dimensional sign change of the gap takes place within this Fermi surface, as shown in Fig. 4. The gap function on the α_1 Fermi surface is schematically shown at the bottom of Fig. 4. This sign change can be considered to be due to the repulsive intraband interaction within the α_1 Fermi surface (shown

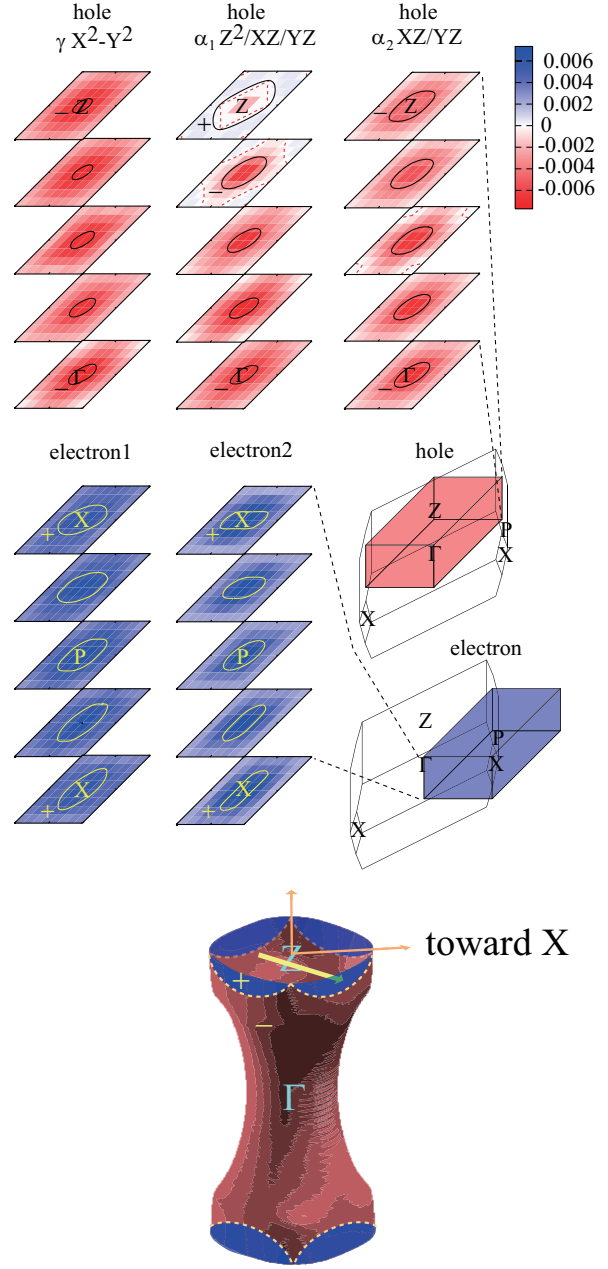


Fig. 4. (Color online) Contour plots of the gap function on the five Fermi surfaces for five horizontal cuts around Γ –Z (hole) or X –P–X (electron). The solid lines represent the Fermi surfaces, while the dashed lines are the nodes of the gap. The Brillouin zone is shown along with the regions where the contour plots of the gap are presented. In the bottom, a schematic view of the gap function on the $\alpha_1 = Z^2/XZ/YZ$ hole Fermi surface is shown. The arrow bridging the Fermi surface indicates the intraband interaction that induces the sign change of the gap.

by the arrow in the schematic figure), which becomes more effective as the volume of the Fermi surface around the Z point grows. In fact, we find that the plus-sign region of the gap on the α_1 Fermi surface tends to shrink for smaller values of x . A similar three-dimensional sign change of the superconducting gap has been found in a five orbital study of Ba122 in ref.³³, although in the model of ref.³³, the α_1 Fermi surface around the Z point has an $X^2 - Y^2$ character (or xy in the notation of ref.³³.) rather than Z^2 .

Since the three-dimensional nodes in the present study occur at portions of the Fermi surface having a strong Z^2 character, the presence of the nodes does not strongly reduce T_c . This is in contrast to that in the case of LaFePO_4 , where nodes of the gap, according to a similar RPA and other theoretical studies, enters in the $X^2 - Y^2$ and/or XZ/YZ Fermi surfaces,^{8, 14–16, 42} which play an important role in spin-fluctuation-mediated superconductivity. The present view explains why T_c is relatively high in $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ despite the presence of nodes in the superconducting gap.

To summarize, we have obtained a three-dimensional ten-orbital model for the isovalent doping material $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$, and applied RPA to obtain the superconducting gap originating from spin-fluctuation-mediated pairing. The obtained gap is basically $s\pm$, and we find no gap nodes on the electron Fermi surface because the $X^2 - Y^2$ γ Fermi surface persists up to a high P content. On the other hand, there are three-dimensional nodes on the $Z^2/XZ/YZ$ portion of the hole Fermi surface. We presume that these three-dimensional nodes in the $Z^2/XZ/YZ$ hole Fermi surface are responsible for the experimentally observed nodal behavior of the superconducting gap of $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$, since the presence of nodes in this material does not seem to strongly affect T_c . The relation between the present view and other experimental results will be of great interest. A comparison between results of neutron scattering experiment and theoretical analysis based on the present gap structure is now in progress.⁴³ It is also necessary to understand how such a gap structure on the hole Fermi surface can be consistent with results of the ARPES experiment,²¹ or the apparently “nodeless” behavior observed in a specific heat experiment.⁴⁴ We note here that our study does not completely rule out the possibility of nodes on the electron Fermi surface, since correlation effects that are not taken into account in the present “band calculation+RPA” study may give rise to gap nodes on the electron Fermi surface (even when the $X^2 - Y^2$ hole Fermi surface is present).¹⁴ Also, there is a possibility that in the *electron*-doped 122 materials, where the γ Fermi surface is less effective, the nodes may be on the electron Fermi surface, as suggested by a recent study.³² In any case, the tendency of three-dimensional nodes entering the $Z^2/XZ/YZ$ hole Fermi surface of the 122 materials should remain as long as this Fermi surface is sufficiently large and the repulsive intraband interaction is effective. In this context, a “heavily hole-doped” material, KFe_2As_2 , mentioned as another nodal superconductor in the introductory part, is also of great interest. In fact, in our preliminary study of this material, we find a similar tendency of three-dimensional gap nodes entering the $Z^2/XZ/YZ$ hole Fermi surface. In this study, we use a model that quantitatively reproduces the spin fluctuation modes observed in a recent neutron scattering experiment.⁴⁵ The details will be published elsewhere.

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